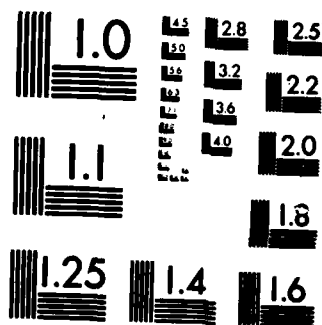


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EFFECT OF LIQUID COMPOSITION ON ENHANCED FLOW DUE TO SURFACE SHEAR IN THE CONTACT LINE REGION: CONSTANT VAPOR PRESSURE BOUNDARY CONDITION

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ABSTRACT

The physicochemical phenomena associated with fluid flow in an evaporating ultrathin film of an ideal binary mixture in the contact line region are discussed. The effect of composition and temperature gradients on surface shear is analyzed and the results are compared with experimental trends. Although simple in concept, the constant vapor pressure boundary condition appears to be very useful in delineating the relative effects of surface tension, temperature and composition on fluid flow due to surface tension gradients in general. (Reprints)

NOMENCLATURE

A_{slv} = Hamaker constant
 \bar{A} = $-A_{slv}/6\pi$
 B = parameter in Equation (3)
 D = intermolecular distance
 g = acceleration due to gravity
 K = curvature
 n = parameter in Equation (3)
 p = pressure
 p_0 = pressure in the vapor space
 p_v = vapor pressure
 p_v^s = saturation vapor pressure
 P = $p + \phi$
 T = absolute temperature
 u = velocity in the x direction

v = velocity in the y direction
 x = distance parallel to the main direction of flow
 x_n = concentration of component n ($n = 1$ or $n = 2$)
 y = distance perpendicular to the main direction of flow

Greek

Γ = mass flow rate
 δ = film thickness
 θ = angle of inclination
 μ = absolute viscosity
 ν = kinematic viscosity
 ρ = density
 σ = surface tension
 τ_{yx} = shear stress in x direction on a surface perpendicular to the y direction
 ϕ = potential energy function per unit volume
 ϕ_B = potential energy per unit volume of a surface molecule of a bulk phase

Subscripts

1 = lighter component in a binary mixture
2 = heavier component in a binary mixture
ls = liquid-solid

ll = liquid
 ss = solid
 slv = solid-liquid-vapor

INTRODUCTION

In a recent set of publications, the use of a scanning microphotometer to help determine the heat transfer characteristics of a thin stationary evaporating film of liquid in the film thickness range $\delta < 10^{-5}$ m was described [1-5]. A simple diagram of the system is presented in Fig. (1). The microphotometer was used to determine the thickness profile by measuring the relative location of interference fringes formed in the film by the reflection of light. Since various mechanisms causing fluid flow are a function of the film profile it can be used to evaluate their effect on evaporative heat transfer. Mixtures of alkanes of various weight percent were studied. The initial studies were concerned with relatively pure systems and were designed to measure the effect of evaporation rate on the thin film profile [1]. Fluid flow resulting from a capillary pressure gradient (due to a curvature gradient) and from a disjoining pressure gradient (due to a thickness gradient) were identified. The evaporating thin films were found to be very stable with fluid flowing towards the heat source. In addition, small percentages of a second component were experimentally found to be very important. It became apparent that even small quantities of an impurity with a substantially lower vapor pressure would concentrate as a result of distillation as the fluid flowed towards the heat source. Subsequent studies demonstrated that small changes in the bulk composition significantly altered the characteristics of the transport processes in the contact line region of evaporating thin films [2-5]. The curvature gradient at the liquid-vapor interface was found to be a strong function of evaporation rate and bulk composition, and to be strongly coupled to the concentration gradient. Concentration and temperature gradients apparently caused interfacial shear stresses and fluid flow patterns that enhanced contact line stability. Although the thickness profile could be easily measured optically, the concentration and temperature profiles in a region less than 10^{-3} m in length could not be easily obtained experimentally. Therefore, extensive modeling of the fluid flow mechanisms dependent on these variables is needed. Herein, a simple model that allows the relative importance of various physicochemical phenomena on fluid flow in an evaporating thin film to be evaluated is presented. The effect of composition and temperature gradients on the surface shear is emphasized. The experimental results on evaporating thin films of mixtures [2-5] parallels in many ways the pioneering experimental results obtained by Bascom, Cottington and Singletary on spontaneous spreading [6]. This research on spreading concerned extremely low evaporation rates. However, surface tension gradients due to concentration gradients were identified as the driving force. This work was extended by Ludviksson and Lightfoot who used a nonisothermal plate with its warm end partially immersed and fluid flowing away from the heat source [7]. We note that there are many technologies in heat transfer that rely on evaporating thin films that are related to the basic problem addressed here.

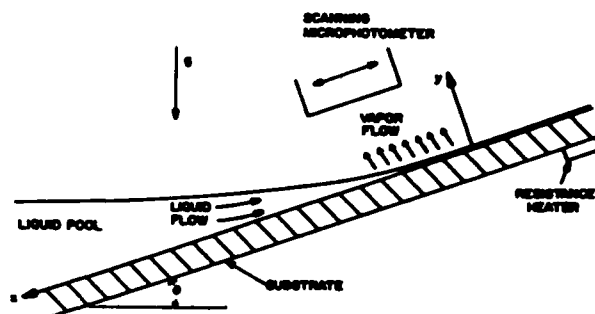


Fig. 1. Evaporating Extended Meniscus on Inclined Flat Plate Immersed in a Pool of Liquid.

FLUID FLOW IN AN EVAPORATING FILM

A continuum model is used to discuss the physicochemical phenomena of importance to fluid flow in a thin film with a varying thickness even though the thickness in a portion of the flow field can be less than the minimum thickness where the continuum model is obviously justified. However, we feel that, as a minimum, the resulting insights justify this presumptive extension to include ultra-thin regions in the initial discussion. We also note that the second order effect of diffusion is neglected in this paper even though the effect of concentration on the surface tension gradient is discussed. Neglecting inertia terms and the vertical component of velocity the following simplified Navier-Stokes equation can be written for the velocity in the evaporating thin film shown in Fig. (1).

$$\mu \frac{\partial^2 u}{\partial y^2} = \frac{\partial}{\partial x} (p + \phi) = \frac{\partial p}{\partial x} \quad (1)$$

In which

$$\phi = -\rho g x \sin \theta + \phi_B - \frac{B}{\delta^n} \quad (2)$$

$$\frac{B}{\delta^n} = -\frac{A_{slv}}{6\pi\delta^3} = \frac{\bar{A}}{\delta^3}, \quad \delta \lesssim 100 \text{ \AA} \quad (3)$$

$$\frac{B}{\delta^n} = \frac{\bar{B}}{\delta^4}, \quad \delta \gtrsim 200 \text{ \AA} \quad (4)$$

Herein we use a potential energy function per unit volume which is a function of the film thickness, $y = \delta(x)$, to represent the difference in behavior of a thin film relative to that of a semi-infinite bulk phase [8-13]. ϕ_B is the potential energy per unit volume of a surface molecule of the bulk phase. Although the results are the same for all practical purposes as those obtained using a pressure jump at the interface, e.g. [14,15], this approach circumvents the need to discuss disjoining pressure expli-

citly. The Hamaker constant for a spreading system is $A_{slv} < 0$. Equation (1) can be solved for the velocity distribution using the following boundary conditions.

$$y = 0, \quad u = 0 \quad (5)$$

$$y = \delta(x), \quad \tau_{yx} = \frac{d\sigma}{dx}, \quad \sigma = \sigma(T, x_n) \quad (6)$$

$$y = \delta(x), \quad P = p_0 + 2\mu \frac{\partial v}{\partial y} + \phi_B - \frac{B}{\delta^n} - \sigma K \quad (7)$$

The letter x without a subscript refers to position, whereas, the letter x with a subscript refers to concentration. The second boundary condition can have a large effect on the velocity distribution because large gradients in the concentration and temperature are possible in evaporating thin films. Even a bulk fluid that is relatively pure can develop large concentration gradients in an evaporating thin film due to distillation. For extremely thin films the surface tension is also a function of the film thickness. These additional effects due to the effect of the film thickness on the surface tension are beyond the scope of the present paper. The total pressure in the vapor space, p_0 , is not equal to the vapor pressure because it includes a variable non-condensable component. We note that the dispersion term gives a bulk effect and the surface tension a surface effect on fluid flow in the thin film. Using Equations (1-7) we obtain Equation (8) for the integration of ρu across the film.

$$r = \frac{\delta^3}{3\nu} [1.5\delta^{-1}\sigma' + \delta^{-n}B' + K\sigma' + \sigma K' - nB\delta^{-(n+1)}\delta'] \quad (I) \quad (II) \quad (III) \quad (IV) \quad (V)$$

$$- \rho g \delta' \cos \theta + \rho g \sin \theta] \quad (8)$$

$$(VI) \quad (VII)$$

The prime refers to differentiation with respect to x . In this paper we will focus on the first three terms in the above brackets which account for flow due to gradients in the intermolecular forces resulting from gradients in the concentration and temperature:

$$(I) = 1.5 \delta^{-1} \sigma' \quad (9)$$

$$(II) = \delta^{-n} B' \quad (10)$$

$$(III) = K\sigma' \quad (11)$$

The fourth and fifth terms primarily depend on the film profile which can be experimentally determined and have been found to be functions of the composition and evaporation rate [1-5]. The final two terms are relatively small except when $r \rightarrow 0$ and can be evaluated. Experimental data have demonstrated that the third term is small compared to the first term since K is usually small compared to $1.5 \delta^{-1}$ [1-5]. At present, the effects of the first two terms on fluid flow are the primary unknowns and will be evaluated below.

COMPARISON OF EFFECTS DUE TO SURFACE AND BULK POTENTIAL GRADIENTS

The following model of Equation (9) is formulated to evaluate the significance and direction of surface tension driven flow for a qualitative understanding of past experimental data. Since the surface tension is a function of both the concentration and the temperature it is advantageous to use an equilibrium boundary condition of a constant vapor pressure for the most volatile component. Physically it seems reasonable to assume that the vapor pressure for a short distance along the vapor-liquid interface would be constant and approximately equal to the equilibrium vapor pressure if the interfacial resistance to evaporation is relatively small and if tangential vapor flow occurs rapidly. In Appendix I, a boundary condition based on effusion is found to be approximately equal to a constant vapor pressure boundary condition for the conditions under study. We note that the constant vapor pressure boundary condition is not a constant evaporative heat flux boundary condition because the variation of composition across the film is not addressed. Therefore, the concept appears to be of general use when discussing the surface shear direction. In addition, the assumption that the second component of the binary mixture being modeled has a negligible vapor pressure is in agreement with some past experimental systems [2-5].

The variation with position of the surface tension of a binary mixture can be written as

$$\frac{d\sigma}{dx} = \frac{\partial \sigma}{\partial T} \frac{dT}{dx} + \sum_n \frac{\partial \sigma}{\partial x_n} \frac{dx_n}{dx} \quad (12)$$

in which the letter x without subscript refers to position.

Using Equation (13) in this equation gives Equation (14) for a binary mixture.

$$\sigma = x_1 \sigma_1(T) + x_2 \sigma_2(T) \quad (13)$$

$$\frac{d\sigma}{dx} = (x_1 \frac{d\sigma_1}{dT} + x_2 \frac{d\sigma_2}{dT}) \frac{dT}{dx} + (\sigma_1 - \sigma_2) \frac{dx_1}{dx} \quad (14)$$

Using Equation (15), which is an extension of Raoult's Law that includes the effect of a "pressure jump" at the liquid-vapor interface, $\Delta P = p_0 - P$, for the variation of vapor pressure with temperature, pressure and composition gives Equation (16) for the variation of vapor pressure with position. The current model applies to that portion of the thin film where the curvature is sufficiently small and the thickness is sufficiently thick so that the effects of curvature and the interfacial force due to the solid on the vapor pressure can be neglected. Therefore, the "pressure jump" at the interface is taken to be zero.

$$p_{v1} = x_1 p_{v1}^s \exp \left(\frac{v_1 \Delta P}{RT} \right) \quad (15)$$

$$\frac{dp_{v1}}{dx} = x_1 \frac{dp_{v1}^s}{dx} + p_{v1}^s \frac{dx_1}{dx} \quad \text{for } \Delta P \rightarrow 0 \quad (16)$$

Imposing a constant component vapor pressure boundary condition gives Equation (17) which can be used along with Equation (14) to give Equation (18) for the variation of surface tension with position.

$$\frac{dx_1}{dx} = -x_1 \frac{d \ln p_{v1}^s}{dx} = -x_1 \frac{d \ln p_{v1}^s}{dT} \frac{dT}{dx} \quad (17)$$

$$\frac{d\sigma}{dx} = \left[(x_1 \frac{d\sigma_1}{dT} + x_2 \frac{d\sigma_2}{dT}) - x_1(\sigma_1 - \sigma_2) \frac{d \ln p_{v1}^s}{dT} \right] \frac{dT}{dx} \quad (18)$$

The evaluation of the various terms in Equation (18) is discussed in Appendix II and the results are presented in Figures (2-4). The particular systems selected were studied in Refs. [2-5]. Before discussing these results, the relative importance of the bulk flow term, Eq. (10), will be analyzed in the next paragraph.

Using Equation (19) for the substrate portion of the liquid-solid Hamaker constant [16] in Equation (3) gives Equation (20).

$$A_{ls} = (A_{ll} A_{ss})^{1/2} \quad (19)$$

$$A_{slv} = A_{ll} - (A_{ll} A_{ss})^{1/2} \quad (20)$$

Taking the derivative of Equation (20) with respect to position while assuming that A_{ss} is not a function of position gives Equation (21).

$$\frac{dA_{slv}}{dx} = \frac{dA_{ll}}{dx} \left[1 - \left(\frac{A_{ss}}{4A_{ll}} \right)^{1/2} \right] \quad (21)$$

Using $A_{ll} = 24\pi D^2 \sigma$ for the liquid film portion of the Hamaker constant [17] in Equation (21) gives

$$\frac{dA_{slv}}{dx} = 24\pi D^2 \left[1 - \left(\frac{A_{ss}}{4A_{ll}} \right)^{1/2} \right] \frac{d\sigma}{dx} \quad (22)$$

The distance D is the order of intermolecular distances. Taking the ratio of Equations (9) and (10) using Equations (3) and (22) gives Equation (23) which demonstrates that the effect due to a variation of surface tension, Equation (9), is usually more significant than the effect due to a variation in the Hamaker constant, Equation (10), since $\delta \gg D$.

$$\frac{(II)}{(I)} = -\frac{8}{3} \left(\frac{D}{\delta} \right)^2 \left[1 - \left(\frac{A_{ss}}{4A_{ll}} \right)^{1/2} \right] \quad (23)$$

We note that the additional effect of thickness on the vapor pressure in Equation (16) still needs to be evaluated for completeness.

DISCUSSION OF RESULTS

The model predicts the following experimentally observed trends [2-5]:

The Formation of a Bulge in the Thin Film Portion of the Extended Meniscus.

The results presented in Figures (2) and (3) demonstrate that the effect of surface shear on fluid

flow changes sign in a distilling thin film as it flows towards the heat source along a constant vapor pressure line with ($\sigma_2 > \sigma_1$). Assuming that the flow starts in a region where surface shear enhances the flow towards the heat source, $d\sigma/dT > 0$, a composition is reached in the flowing film at which flow reversal can occur in the film. Assuming further that there is an initial transient period during which the flow at this point is greater than the evaporation rate above it, a liquid reservoir in the shape of a bulge should form since some evaporation in this region would lead to a concentration at a higher temperature closer to the heat source at which the surface shear opposes the flow. This result appears to be of general importance to the formation and movement of lenses. The results presented in [2-5] and in the figures demonstrate that the formation of a bulge is a function of the bulk concentration, temperature level and $\Delta\sigma$: an increase in the concentration of a lower boiler with a significantly different surface tension in the bulk leads to an increase in the propensity to form a pool above the meniscus for a limited range of the surface temperature and concentration. We note that the coupling between the initial composition, the fraction distilled and the final composition is not presented in the figures.

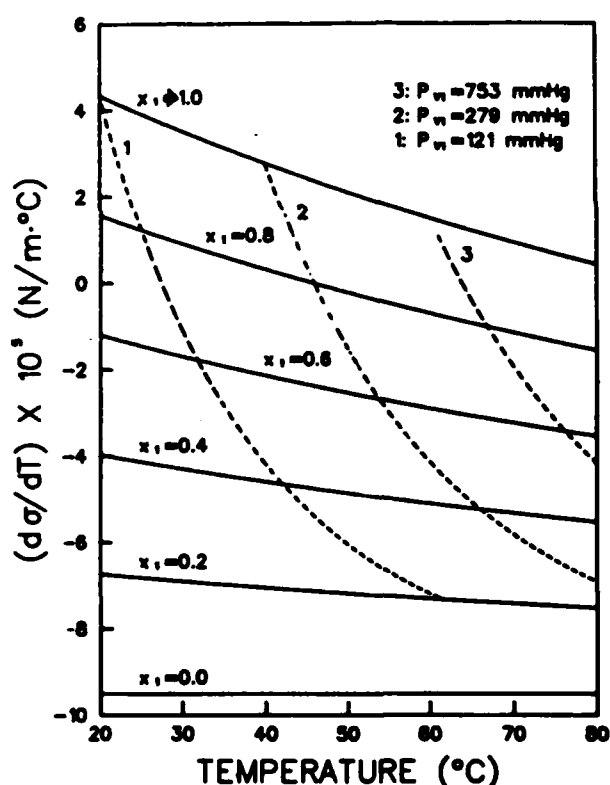


Fig. 2. Surface Tension Derivative (Coefficient in Equation (18)) Versus Temperature for Hexane-Octane Mixtures.

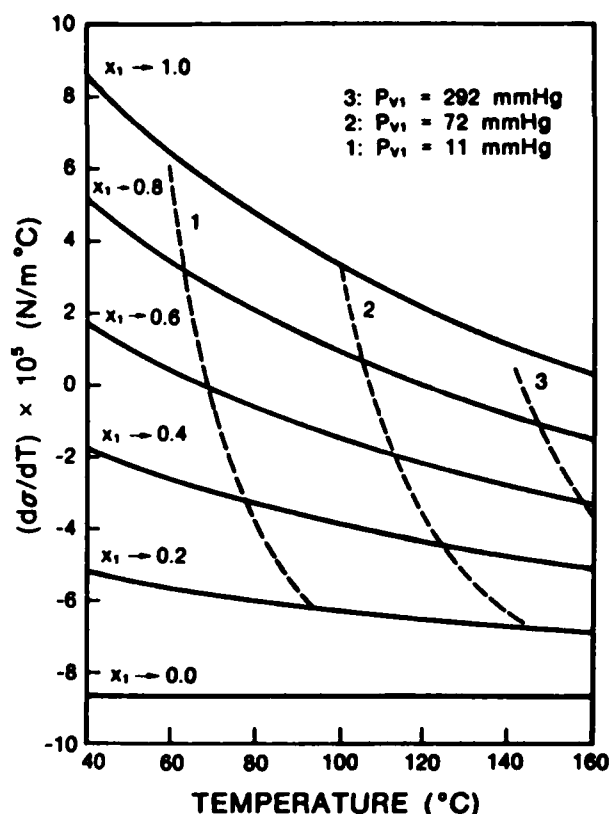


Fig. 3. Surface Tension Derivative (Coefficient in Equation (18)) Versus Temperature for Decane-Tetradecane Mixtures.

The Formation of a Bulge would not be Observed with a Very Pure Fluid.

Since distillation would change the composition of a very pure fluid very slowly, this case is represented by $x_1 = 1.0$ in the figures. The results presented in Figure (4) demonstrate that the surface shear is away from the higher temperature if the difference in the surface tensions, $\Delta\sigma = (\sigma_1 - \sigma_2)$, is relatively small, which is the usual case for relatively pure samples. In these systems flow towards the heat source is due to Terms (IV & V) in Equation (8). We note that small residual lenses were observed in [1].

The Curvature Gradient is a Strong Function of the Bulk Composition.

Equation (8) demonstrates that the curvature gradient and the surface tension gradient are strongly coupled. Therefore, a change in the surface tension gradient for the same mass flow rate could appear as a change in the curvature gradient in the reported experimental results. When the experimental results were analyzed in Ref. [2-5] using only Terms (IV - VII) in Equation (8), relatively more flow apparently occurred with the pure system than with the mixture for the same heat flux based on the temperature measurements. This apparent difference could be the result of the coupling between the various terms in Eq. (8).

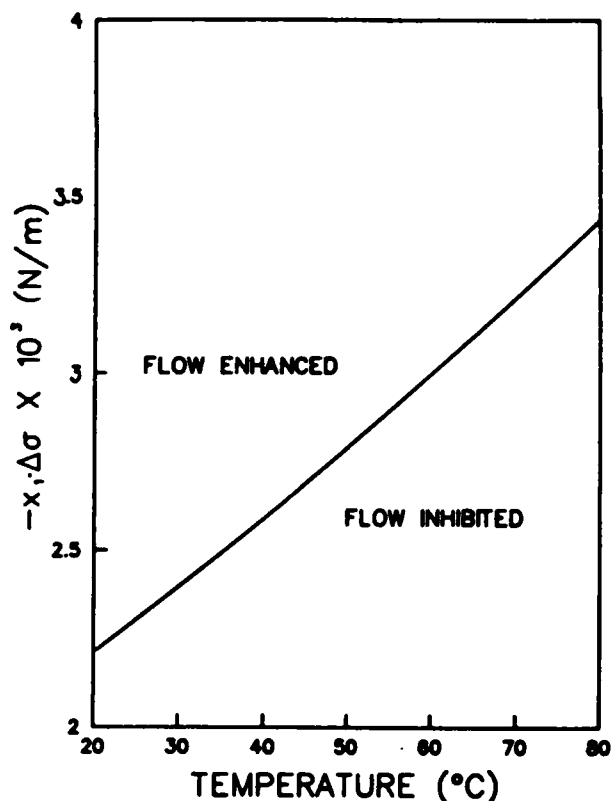


Fig. 4. $x_1 \Delta\sigma$ Versus Temperature for $\sigma' = 0$ and

$$\frac{d\sigma_1}{dT} = \frac{d\sigma_2}{dT} = -10^{-4} \text{ N/mK for Hexane.}$$

CONCLUSIONS

A simple model based on continuum mechanics, interfacial phenomena, a constant vapor pressure boundary condition, and an extension of Raoult's Law qualitatively describes the following trends experimentally observed in experiments of fluid flow in evaporating thin films in the contact line region:

- 1) The effect of surface shear can change sign in a distilling thin film as it flows towards the heat source along a constant vapor pressure line;
- 2) There is a significant range of composition and temperature over which surface shear enhances flow towards the heat source;
- 3) Surface shear inhibits flow towards the heat source in a relatively pure fluid when the difference in component surface tensions is small; and
- 4) The surface tension gradient and the curvature gradient are strongly coupled.

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APPENDIX I

Schrage [18] used the concept of effusion to derive an equation for the net mass flux of matter crossing a liquid-vapor interface which has been used extensively, e.g., [13,19-21]. The mass flux due to effusion at an interface is proportional to p_v/T . Taking this to be constant at the liquid-vapor interface an alternate equation to Equation (18) can be derived for the variation of surface tension with position.

Using

$$p_{v1} = x_1 p_{v1}^S T^{-1/2} \quad (A-1)$$

in place of Eq. (15) gives

$$\frac{dx_1}{dx} = -x_1 \left[\frac{d \ln p_1^S}{dT} - \frac{1}{2T} \right] \frac{dT}{dx} \quad (A-2)$$

For temperatures of interest herein, the second term in the brackets in Eq. (A-2) is small compared to the first term.

APPENDIX II

In order to evaluate the surface tension as a function of temperature the following equations recommended by Jasper [22] were used (Temperatures, T_c , are in Celsius units):

Hexane: $\sigma = (20.44 - 0.1022 T_c) \times 10^{-3} \text{ N/m}$

Octane: $\sigma = (23.52 - 0.09509 T_c) \times 10^{-3} \text{ N/m}$

Decane: $\sigma = (25.67 - 0.09197 T_c) \times 10^{-3} \text{ N/m}$

Tetradecane: $\sigma = (28.30 - 0.08682 T_c) \times 10^{-3} \text{ N/m}$

The following Antoine equation was used to represent the saturation vapor pressure in mmHg for the alkanes:

$$\ln p_v^S = A - \frac{B}{T_c + C} \quad (A-3)$$

Differentiation of Eq. (A-3) leads to

$$\frac{d \ln p_v^S}{dT} = \frac{B}{(T_c + C)^2} \quad (A-4)$$

The following Antoine constants were found in the literature:

| | A | B | C |
|-------------|---------|---------|--------|
| Decane [23] | 16.0114 | 3456.80 | 194.48 |
| Hexane [23] | 15.8366 | 2697.55 | 224.37 |

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